Radiation heat savings in polysilicon production: Validation of results through a CVD laboratory prototype

A. Ramos , C. del Cañizo, J. Valdehita, J.C. Zamorano, A. Luque

A B S T R A C T

This work aims at a deeper understanding of the energy loss phenomenon in polysilicon production reactors by the so-called Siemens process. Contributions to the energy consumption of the polysilicon deposition step are studied in this paper, focusing on the radiation heat loss phenomenon. A theoretical model for radiation heat loss calculations is experimentally validated with the help of a laboratory CVD prototype. Following the results of the model, relevant parameters that directly affect the amount of radiation heat losses are put forward. Numerical results of the model applied to a state-of-the-art industrial reactor show the influence of these parameters on energy consumption due to radiation per kilogram of silicon produced; the radiation heat loss can be reduced by 3.8% when the reactor inner wall radius is reduced from 0.78 to 0.70 m, by 25% when the wall emissivity is reduced from 0.5 to 0.3, and by 12% when the final rod diameter is increased from 12 to 15 cm.

1. Introduction

In the last years the polysilicon market has experienced a turbulent stage in which Photovoltaics (PV) has by far surpassed Microelectronics as the main consumer [1]. Driven by the exponential growth of PV, first the industry suffered a silicon shortage. Once the efforts of many players gave place to new capacity being installed, the scenario quickly changed to that of oversupply. Prices have fallen dramatically, putting pressure on those who cannot deliver the most competitive technology. Notwithstanding, the growth foreseen for PV in the future implies the need for new capacity in the medium term and polysilicon prices are forecasted to become stable in the next years [2,3].

In this context, it is important for polysilicon producers to reduce production costs and thereby allow themselves to make their way into polysilicon production tier 1. Furthermore, the polysilicon cost impacts significantly on the total cost of PV and on the energy payback time [4,5]; this feedstock is responsible for one-third and one-fourth of the final PV module cost and for 28% of the total energy payback time [6]. Due to the fact that specifications for PV polysilicon, called solar grade silicon (SoG Si), can be relaxed as compared to those for microelectronics, process simplifications and innovations can be implemented, leading to cost reductions.

The currently favoured means of obtaining polycrystalline SoG Si is via the chemical route, which leads to high quality polysilicon.

The chemical route, in particular the so-called Siemens process, consists in polysilicon deposition by chemical vapour deposition (CVD) from trichlorosilane (TCS) [7]. It comprises first the transformation from the metallurgical silicon (MG Si) to TCS and other by-products, and second the TCS decomposition into high purity silicon. Overall energy consumption for a polysilicon plant has been reduced in the last years, from around 90–140 kWh/kg for large capacity plants [8,9] to something in the range of 70 kWh/kg for last generation plants [10]. The largest contributor towards this is the second step (the CVD process itself), with an estimated consumption in the best case in the range of 45–50 kWh/kg. Several solutions are proposed to reduce this high energy use, such as recycling gaseous by-products or heat recovery.

This paper is centred in the second step of the Siemens process: the polysilicon CVD. It presents empirical results with a laboratory scale Siemens prototype through which we validate a theoretical radiation heat loss model developed in our institute. Thus, on the basis of this model, the key parameters involved in radiation heat savings are identified.

2. Important factors for energy consumption

A deep knowledge of the CVD process involves jointly studying polysilicon deposition conditions (fluid dynamic theory), radiation heat emitted by the hot silicon rods (radiation heat transfer theory), electrical heating of the rods (electromagnetic theory) and chemical reactions.

In industry, the TCS decomposition or CVD of polysilicon is conducted in a reactor chamber of 10–16 m^3. Inside the reactor chamber, a number of inverse U-shape polysilicon rods are placed. These U-shape rods are heated by the Joule effect to around 1100 °C in a Hydrogen and TCS atmosphere. A surface reaction occurs resulting in high purity silicon deposition, which causes the U-shape rods' diameter to increase with time.

The typical batch time of a CVD process in industry is 80–100 h divided in three steps: heating, deposition and cooling down. Almost all the energy is consumed during the deposition step: cooling down requires negligible energy consumption and the heating time is typically less than 1 h.

Contributions to the high energy consumption of the polysilicon CVD are heat loss due to radiation, convection, thermal conduction and heat consumed due to the chemical reactions taking place. Of these, the major contributors are radiation and convection heat loss.

Focusing on the deposition step, the polysilicon deposition rate and hence the growth of the rod diameter is very slow: in the order of microns per minute. Thus, at each instant in the process, the system can be considered to be in the steady state. Then, the power supplied instantly must compensate for radiation and convection heat loss and the heat consumed by the chemical reaction.

Radiation heat loss for industrial Siemens reactors becomes the greatest percentage of the total energy consumed: 60–75% (assuming that all the energy radiated by the rods towards the reactor wall is wasted). Convection heat loss is between 23.5 and 38.5% and the heat consumption due to the chemical reaction sums up to 1.5% [11]. These figures indicate the importance of a reliable tool allowing research on radiation heat savings.

2.1. Theoretical model for radiation heat loss

A theoretical model for radiation heat loss calculations has been developed in our institute. It was presented and described in detail in [12], and it has been applied to three different reactor configurations in [13]. Here, the theoretical model is briefly reviewed.

The energy radiated by the rods towards the reactor wall is mainly dependent on the *emissivity* of the rods and the wall, their corresponding *temperature* and, finally, on the geometries and the geometrical arrangement of the rods inside the reactor chamber.

In addition, the rate of outgoing radiant heat per unit area from the rods, called *radiosity*, is the sum of the *directly emitted radiation heat flux* and the reflected portion of the *incoming radiant heat flux* (which is determined by the surface *reflectivity*). This is sketched in Fig. 1. The fraction of radiative heat flux leaving a certain surface that arrives to another is determined by the geometrical factors, which are defined in [14].

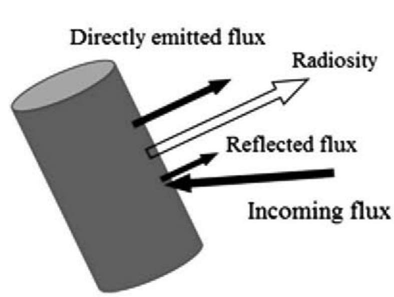

Fig. 1. Outgoing radiant heat per unit area from the surface (radiosity).

Once the material properties, the geometrical arrangement, the surface temperatures and the incoming and directly emitted radiant heat flux are known, the *net heat flux exchanged* by a certain surface area in watts is calculated. This corresponds to the radiation heat loss by this surface.

For a single rod placed in the reactor chamber, all heat radiated from the rod arrives at the reactor wall which is maintained at a constant temperature due to water cooling; it absorbs the radiation heat partially and some small part is reflected back. The wall also directly emits radiation heat flux, although this is much lower than the rods due to the material and the temperature. This small amount, plus the reflected part from the incoming flux, not only reaches the rod surface but also the reactor inner wall. In addition, as the diameter of the rod increases, a higher percentage of the radiosity of the wall reaches the rod; therefore, the radiation heat loss per unit area decreases throughout the deposition process.

For configurations with more than one rod, each rod is additionally heated by the incoming radiosity from the other rods. Both a higher number of rods in the reactor chamber and a more compact arrangement lead to lower radiation heat loss. Again, for these configurations, as the diameter of the rods increases, the radiation heat loss per unit area does the opposite. This tendency is steeper as the number of rods considered and the compactness grow.

3. Research through a laboratory scale reactor

Research on SoG Si through the chemical route using an industrial scale CVD reactor is very costly in time and money. Thus, a laboratory scale reactor allows this research to be carried out in a more affordable way, provided extrapolation of results to the industrial scale can be clearly justified.

At the *Instituto de Energía Solar* (IES), there is an in-house-made CVD prototype with which all working conditions (except the pressure inside the reactor chamber) of the industrial Siemens process can be reproduced. The prototype reactor admits a small number of single silicon rods instead of the typical U-rod shape used at the industry scale. The reactor's chamber is a stainless steel cylinder—upright placed; gases are introduced from the bottom of the reactor's chamber and the outlet is placed at the top. The rods are electrically connected – series connection – to two copper electrodes placed at the bottom and top of the reactor chamber. In Fig. 2 a sketch of the prototype is shown. Rods' temperature is monitored by a two-color pyrometer through a sapphire peephole placed at middle-height of the rods. The temperature is considered to be homogeneous in length considering that the rods' diameter and boundary conditions in length are homogeneous. Gas temperature is measured at the inlet and outlet of the reactor's chamber by thermocouples. Also, the temperature of the inside wall is measured by thermocouples. All the system is water cooled. Last, inlet and outlet gas composition is analysed by means of mass spectrometer. Deposition processes with the prototype are conducted at a constant pressure of 1 bar, around 6 times lower than the typical industrial value. Equations that link together the pressure inside the reactor chamber and the main parameters involved in the process are known, making the extrapolation straightforward [15].

From the experiments conducted with the prototype reactor, in particular for a 9 h deposition process on a single rod at 1150 °C, the percentage of the total energy consumption that corresponds to the heating is 3.2%. The remaining 96.8% is consumed during the deposition step. The longer the deposition process, the longer the deposition step relative to the heating step. Scaling-up to an industrial Siemens process, the energy consumed during the heating step can be disregarded. In any case, we would expect

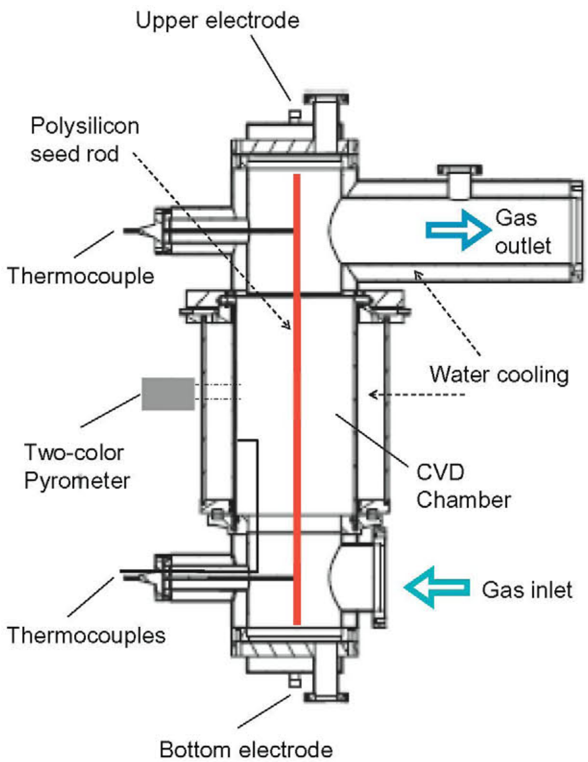

Fig. 2. CVD laboratory prototype at the IES.

any improvement in radiation heat loss during the deposition step to also reduce heat loss in the heating step.

Radiation heat loss for the laboratory scale reactor is responsible for around half of the total energy consumed. For example, for a 9 h deposition process conducted at 1150 °C, radiation heat loss is responsible for 47% of the total energy consumption, convection heat loss for 52.5%, and the chemical reaction for barely 0.5%.

The corresponding percentages for industrial scale reactors – shown in the previous section – will change due to the pressure effect. The convection heat loss is directly related with this parameter; the higher the pressure inside the reactor chamber, the lower the convection heat loss. Therefore, for the extrapolation of the percentages of radiation and convection heat loss from the laboratory scale to the industrial one, process pressure differences must be considered.

4. Experimental results for model validation

Two different configurations with one and four rods were reproduced with the prototype reactor to validate the theoretical model.

In the experiments conducted with the prototype, the power consumption is measured throughout the process. The following are monitored: the temperature of the rods and the wall, inlet and outlet gas composition, the inlet and outlet gas temperatures and the temperature of the gases inside the reactor chamber. The only variables that change appreciably during the process are the surface temperature of the rods and the temperature of the gases inside the reactor chamber. The former greatly affects the power consumption and the latter increases with time due to the increase of the radiating area. Therefore, for the results hereunder presented, the real values of the two most relevant parameters: the temperature of the rods' surface and the gasses, are considered. The reactor wall and rods' emissivities are known and equal to 0.5 and 0.7, respectively. Besides, all numerical results correspond to the particular CVD prototype geometry.

Convection heat loss can be calculated from experimental data after the deposition process ends. The power lost instantly by convection depends on inlet and outlet gas temperatures, the heat capacity at constant pressure of the gases and the mass exchange between the inlet and outlet. The power consumed due to the chemical reactions taking place is calculated knowing the reaction enthalpy, the solid silicon density and the silicon growth rate [11]. The total power consumption is known by means of the power supplied by a three-phase transformer.

The chemical reaction and convection heat loss calculated are subtracted from the measured power consumption, resulting in the radiation heat loss. For the two different configurations, this subtraction is compared with the theoretical calculations for radiation heat loss.

In Fig. 3, the different configurations reproduced in the laboratory reactor are sketched. Due to functional requirements, the two configurations differ in both the length of the rods and their initial and final diameters. This does not present a problem, since the purpose of this experiment is to validate the model and not to make comparisons between different configurations.

4.1. Single rod configuration

The crosses in Fig. 4 show the measured radiation heat loss as a function of the rod diameter over the course of a single experiment. The solid line shows the respective values predicted using the model. The deposition time shown is around 5 h, the initial rod

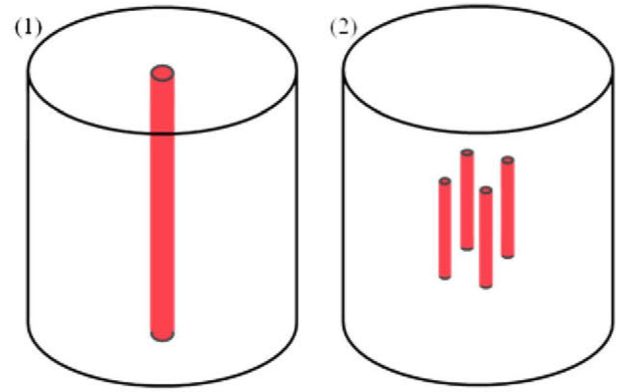

Fig. 3. Configurations reproduced with the CVD prototype. One single rod (1) and four rod (2) configurations.

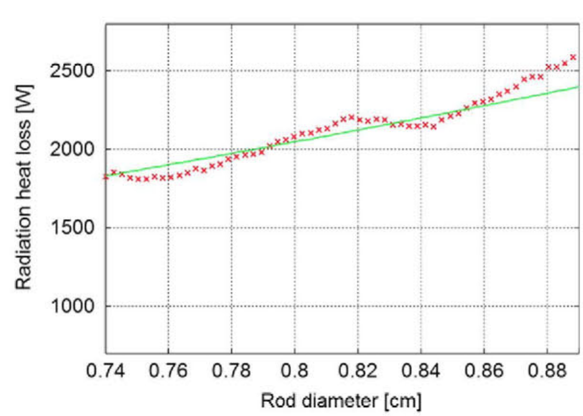

Fig. 4. Radiation heat loss measured (crosses) and theoretical (solid line) for one single rod.

diameter is 0.74 cm, the length of the rod is 53 cm and the temperature of the rod surface is kept between 1130 and 1180 °C.

Good agreement is observed between the measured and predicted values. The power supplied changes in short steps and not steadily because of the control algorithm, provoking the oscillations that can be observed in Fig. 4, which correspond to temperature oscillations in the rod. These oscillations are in the worst case under 2.5%. In terms of power per unit area, the maximum difference between the theoretical calculations and the experimental measurement is 8.3%. The difference between the values averaged over the deposition process is under 1%. The average radiation heat loss per unit area measured is $1.564 \times 10^5\ W/m^2$ and the theoretical model predicts $1.551 \times 10^5\ W/m^2$: a difference of 0.5%.

4.2. Four rod configuration

In Fig. 5, the measured and predicted results for the four rod configuration are presented as before. The deposition time is around 7 h and the temperature of the rods' surface is kept between 1145 and 1185 °C. No deviation in temperature is expected between the four rods due to the symmetry of the arrangement. The initial diameter of the rods is 0.17 cm and the length of the rods is 12 cm; notice that both dimensions are lower than in the previous experiment. Depending on the rods' position inside the reactor chamber, power loss through radiation changes [13]. In this case each rod is placed at the corner of a 2×6 cm side rectangle. This disposition is chosen due to electrical needs but if one disposition with shorter- and longer-distance between the rods was selected, the radiation power loss would decrease or increase, consequently with the change of the geometrical factors.

For this configuration, both curves represented in Fig. 5 are again quite close. The maximum differences between the theoretical calculations and the experimental ones are 16.3%. The difference between the values averaged over the deposition process is about 3.8%. The average measured radiation heat loss per unit area along the deposition process is $1.731 \times 10^5\ W/m^2$, compared to $1.668 \times 10^5\ W/m^2$ for the theoretical model: a 3.6% difference.

5. Discussion

From all the results presented above, the theoretical model can be considered empirically validated. For both configurations, deviation between measured and theoretical results is under the measurement error.

For the model validation through the laboratory scale prototype it is not necessary to reach the relevant rods' diameters at industry because the accuracy of the theoretical model does not change with the rods' diameter. An experiment of several hours is sufficient to see that the diameter growth adheres to the trend of the theoretical model.

Experimental measurements are in watts per square meter – W/m^2– thus, we compare those numbers for two different configurations and the theoretical model results. Absolute numbers as kilowatts-hour per kilogram of silicon – kWh/kg – obtained with the laboratory prototype are several times higher than those at the industry due to the bad compactness of the configuration design (it is not possible to improve it due to electrical power needs, among others) and the process pressure, which is 6 times lower than in industry.

6. Extension to industrial scale reactors

In this section, the validated model is used to predict the radiation heat loss for industrial scale reactors. Different configurations and parameter variations are explored. We consider a typical 36 rod industrial Siemens reactor with a fixed geometrical arrangement and investigate separately the effect of changing the wall emissivity, the reactor inner wall radius and the rod surface temperature. Numerical results for a 36 rod industrial reactor shown correspond to a Siemens reactor at our disposal for research purposes in a near future.

In Fig. 6, the radiation heat loss per rod for different wall emissivities (ε_w) is represented. The reactor's wall is made of stainless steel, whose emissivity is around 0.5. Due to process contamination or previous polishing, reactor's wall emissivities of 0.7 and 0.3 can be measured. Reducing the emissivity of the wall from 0.7 to 0.5, the radiation heat loss decreases by 20%; this reduction is 45% for a wall emissivity of 0.3. Absolute specific numbers as kWh/kg of silicon are related with the polysilicon growth rate, which depends on rods' temperature, pressure and inlet gas composition among other parameters. Fixing a growth rate of 8 μm/min the energy consumed due to radiation is 25.6, 38.0 and 47.9 kWh/kg for wall emissivities of 0.3, 0.5 and 0.7, respectively. Likewise, for a fixed growth rate of 12 μm/min the energy consumed is 18.6, 25.4 and 34.9 kWh/kg.

A similar but less pronounced effect occurs when the reactor wall is placed closer to or further from the rods. In Fig. 7, the radiation heat loss per rod for different wall radii (R_w) is shown. For the curves presented below, the emissivity of the reactor wall is considered constant and equal to 0.5.

If the reactor inner wall radius is displaced from 0.78 to 0.74 m, the radiation heat loss decreases by 1.8%; when the wall radius is 0.70 m this reduction becomes 3.8%. Fixing a growth rate of 8 μm/min the energy consumed is 37.1, 38.0 and 38.6 kWh/kg for inner wall radii of 0.70, 0.74 and 0.78 m, respectively. Also, for a

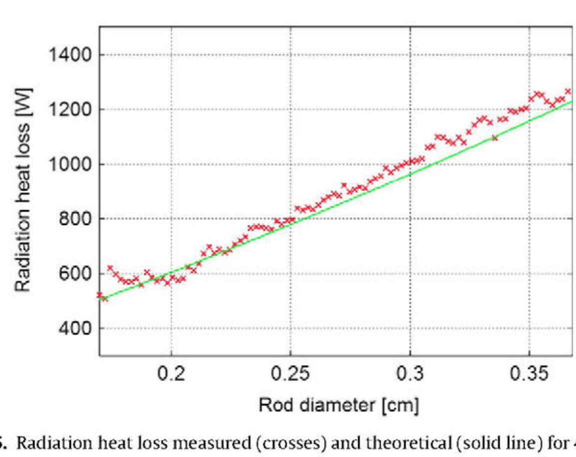

Fig. 5. Radiation heat loss measured (crosses) and theoretical (solid line) for 4 rods.

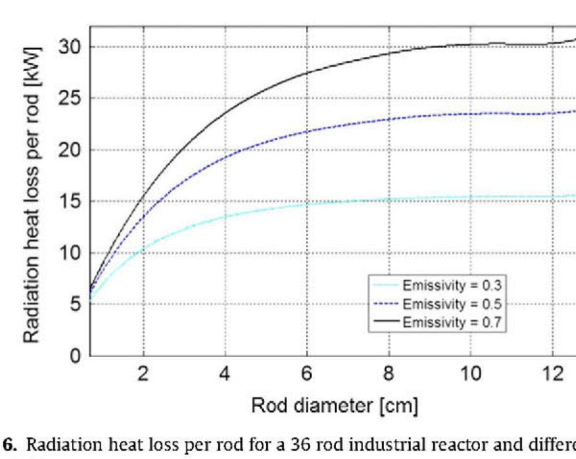

Fig. 6. Radiation heat loss per rod for a 36 rod industrial reactor and different wall emissivities.

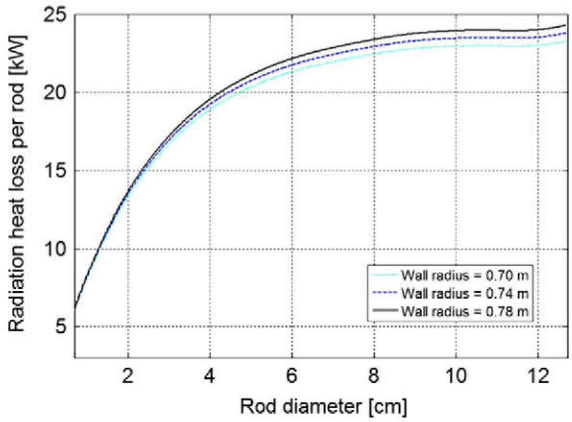

Fig. 7. Radiation heat loss per rod for a 36 rod industrial reactor and different wall radii.

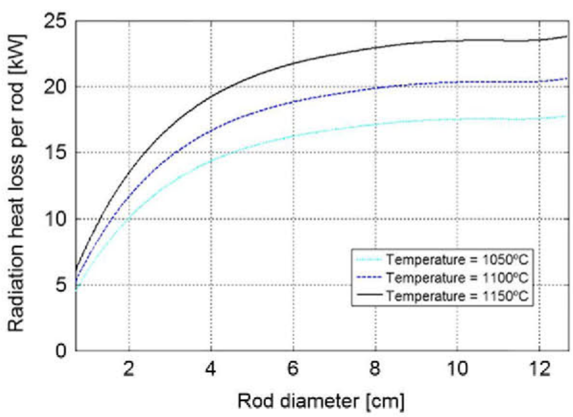

Fig. 8. Radiation heat loss per rod for a 36 rod industrial reactor and different rod surface temperatures.

fixed growth rate of 12 μm/min the energy consumed is 25.4, 27.0 and 28.1 kWh/kg.

So far in this section, all the curves presented consider a constant rod surface temperature of 1150 °C. Higher rods' temperature leads to higher deposition rates, and at the same time higher temperature leads to higher energy consumption. In industry is typical an average deposition temperature of the rods surface around 1100 °C, while the theoretical temperature that minimises the kWh/kg of Si is around 1150 °C. The reason to consider more conservative temperatures is to avoid the appearance of what is known as popcorn [16].

In a typical industrial reactor, the deviation in temperature between the different rods is minimised by controlling the power supplied to different concentric rings of rods separately [7]. Here we assume that all rods have the same temperature. In Fig. 8, for a constant wall emissivity of 0.5 and wall radius of 0.74 m, the radiation heat loss per rod for different rod surface temperatures (T_{rod}) is shown. Decreasing the rod surface temperature from 1150 to 1100 °C, the radiation heat loss decreases by 13.4%; this reduction is 25.4% when the rod surface temperature is 1050 °C. Fixing a growth rate of 8 μm/min, the energy consumed is 28.3, 32.8 and 38.0 kWh/kg for rods' temperatures of 1050, 1100 and 1150 °C, respectively. For a fixed growth rate of 12 μm/min the energy consumed is 20.3, 23.9 and 25.4 kWh/kg.

For all configurations presented in this paper, as the rod diameter increases the radiation heat loss per unit area decreases. Consequently, in energy terms, a CVD process with higher initial and final rod diameters (higher compactness) is cheaper. However, this requires substantially more raw material and also leads to wastage of the production capacity of each reactor. For example, increasing the initial rod diameter from 0.7 to 4 cm reduces the Si production by 10%, but also reduces the radiation heat loss by 45%. When the initial rod diameter is increased to 7 cm the Si production is reduced by 30%, and the radiation heat loss is reduced by 70%. However, the bigger initial rod radius implies that the material at the beginning of each run is 33 times greater in the first case, 100 times greater in the second.

The same phenomenon leads to lower radiation heat loss if the initial rod diameter is kept constant but the final rod diameter is increased. If the deposition process ends when the rod diameter is 15 cm instead of 12 cm, the radiation heat loss is increased by 4.2%, but the silicon production is increased by 56%.

Prior to its validation, this model was applied to calculation of the radiation heat losses for three state-of-the-art industrial size reactor configurations [12]. The amount of radiation heat loss per rod during a CVD process for the case of 48 rod reactor was 10% lower than in the case of 36 rod reactor, and 20% in the case of 60 rod reactor. These previous results are consistent with the ones presented in this work: the radiation heat loss savings are obtained due to the increase of the reactor compactness. Differences between the industrial scale results, for the 36 rods reactor presented in this paper and the former ones, are due to differences in compactness of both reactors. In Ref. [12] the base plate of the 36 rods reactor considered allows a final rod diameter of 18–20 cm while for the present case, the maximum rod diameter of the rods is barely above 13 cm. Furthermore, the former paper presented the strong influence that the wall emissivity has on the radiative power loss, but does not investigate other important parameters for a fixed industrial reactor, as discussed above in this section.

7. Conclusion

In this paper, experimental and theoretical work on the basis of a theoretical model for radiation heat loss is presented. Through a CVD laboratory scale prototype, this radiation model is validated. The model arises as an important tool to deepen understanding of the radiation heat loss phenomenon.

Numerical results for a 36 rod industrial Siemens reactor are shown. Design compactness and wall material properties clearly influence the radiation heat loss. On this line, the highest possible compactness and materials with low emissivities – high reflectivities – are desired. Nevertheless, to look for low emissivity materials to be used as the reactor wall, not only structural constrains exist, but also polysilicon contamination susceptibility must be considered.

In addition, heat loss through radiation is higher for higher rod surface temperatures. A typical value for industrial processes is average rod surface temperatures around 1100 °C. This value is chosen as a compromise between a higher polysilicon deposition ratio and lower power consumption. Higher temperatures lead to faster growth of the rod diameter. Thus, decreasing the rod temperature during the deposition, although lowering the power consumption, lengthens the CVD process. The target figure is to minimise the final energy consumed per kilogram of polysilicon produced.

In short, thanks to this model, it has been put forward that radiation heat loss has a great dependence on the rod's surface temperature. Moreover, the key to diminishing the radiation heat loss, for a fixed rod temperature, is to decrease the reactor wall emissivities and to increase the design compactness.

Acknowledgement

The Spanish *Ministerio de Economía y Competitividad* is greatly acknowledged for its support through project IPT 2012-0340-120000.

Alexander Mellor is acknowledged for his contribution to the manuscript revision.

References

[1] K. Hesse, E. Schindlbeck, E. Dornberger, M. Fischer, Status and development of solar-grade silicon feedstock, in: 24th European Photovoltaic Solar Energy Conference, 2009, pp. 883–885.
[2] M. Meyers, Silicon 2012: the new Si game, in: 10th Solar Silicon Conference, 2012.
[3] No End in Sight for Polysilicon Oversupply, Technical Report, NPD Solarbuzz, 2012.
[4] S. Singer, Photovoltaics: Getting Cheaper, Eco Queen of Green, 2007.
[5] B. Ceccaroli, O. Lohne, Solar grade silicon feedstock, in: Handbook of Photovoltaic Science and Engineering, 2nd ed., 2011, pp. 169–217.
[6] E. Alsema, M. de Wild-scholten, Reduction of the environmental impacts in crystalline silicon module manufacturing, in: 22nd European Photovoltaic Solar Energy Conference and Exhibition, 2007, pp. 829–836.
[7] R.B. Herring, L.P. Hunt, Polysilicon preparation, in: W.C. O'Mara (Ed.), Handbook of Semiconductor Silicon Technology, 1990, pp. 33–93.
[8] Y. Delanoy, Purification of silicon for photovoltaic applications, Journal of Crystal Growth 360 (2012) 61–67.
[9] J. Maurits, Reducing Polysilicon Materials Cost, 13th ed., Photovoltaics International, 2011.
[10] D. Keck, Lowering the cost to produce polycrystalline silicon, in: 10th Solar Silicon Conference, 2012.
[11] G. del Coso, I. Tobías, C. Ca nizo, A. Luque, Temperature homogeneity of polysilicon rods in a Siemens reactor, Journal of Crystal Growth 299 (2007) 165–170.
[12] G. del Coso, C. Ca nizo, A. Luque, Radiative energy loss in a polysilicon CVD reactor, Solar Energy Materials & Solar Cells 95 (2011) 1042–1049.
[13] A. Ramos, C. del Cañizo, J. Valdehita, J.C. Zamorano, A. Rodríguez, A. Luque, On track for Solar Grade Silicon through a Siemens process-type laboratory reactor: operating conditions and energy savings, in: Silicon for the Chemical and Solar Industry XI Conference, 2012.
[14] R. Siegel, J.R. Howell, Thermal Radiation Heat Transfer, McGraw Hill, 1972.
[15] G. del Coso, Chemical Decomposition of Silanes for the Production of Solar Grade Silicon, Ph.D. Thesis, Universidad Politécnica de Madrid ETSIT, 2010.
[16] A. Ramos, C. del Ca nizo, J. Valdehita, J.C. Zamorano, A. Rodríguez, A. Luque, Exploring polysilicon deposition conditions through a laboratory CVD prototype, Physica Status Solidi C 10–11 (2012) 2164–2168.